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Partitioning of Metals in Rotary Kiln Incineration

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Cadmium, lead and bismuth appeared volatile over the range of kiln temperatures tested, while the other six metals displayed refractory properties. Of the three independent variables tested, feed chlorine content had the strongest effect on changes in metal partitioning across the tests; as chlorine content increased, metal volatilization appeared to increase, while scrubber efficiency for metals decreased.

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Cadmium, lead and bismuth appeared volatile over the range of kiln temperatures tested, while the other six metals displayed refractory properties. Of the three independent variables tested, feed chlorine content had the strongest effect on changes in metal partitioning across the tests; as chlorine content increased, metal volatilization appeared to increase, while scrubber efficiency for metals decreased.

INTRODUCTION

Metals, such as arsenic, barium, beryllium, cadmium, chromium, lead, mercury, nickel, and zinc are of concern in waste incineration because of their presence in many hazardous wastes and because of possible adverse health effects from human exposure to emissions. Incineration will change the form of metal fractions in waste streams, but it will not destroy the metals. As a result, metals are expected to emerge from the combustion zone essentially in the same total quantity as the input. The principal environmental concern centers around where and in what physical or chemical form the metals end up in the combustion system, i.e., bottom ash, air pollution control device (APCD) residues, or stack emissions.

Most interest has traditionally focused on stack emissions of metals. However, increasing attention is being given to the quality of residuals from incineration of metal-bearing wastes since disposal of these materials may be subject to restrictions on land disposal under

the Hazardous and Solid Waste Act of 1984 (HSWA). (1)

A major role of the U.S. EPA Combustion Research Facility (CRF) in Jefferson, Arkansas is to perform research in support of regulatory development by the U.S. EPA Office of Solid Waste (OSW). Accordingly, the CRF has conducted research to study the partitioning of trace metals during the incineration of a metal-bearing surrogate waste. (2)

The testing described herein took place in the Rotary Kiln System (RKS) of the CRF, utilizing venturi- and packed tower-scrubbing for particulate- and acid gas-control. This research is part of a larger test program designed to evaluate the fate of metals in incineration using a variety of APCDs.

PURPOSE

Field studies to date indicate that emissions of metals from incinerators burning wastes of relatively low metal content probably do not pose an unacceptable level of risk. However, as the level of metals in wastes which are incinerated rises, there is concern that some incinerators could create unacceptable risk. Current regulatory thinking suggests implementation of metal emission limits and metal feed-rate limits, calculated using health risk levels and dispersion modeling.

Information gathered from the subject research will be used to further examine the impact of metals upon environmental emissions and to assist in developing and refining regulatory strategies for dealing with such an impact. (3)

The research was designed to identify:

- the distribution of metal emissions among ash, scrubber blowdown water, flue gas particulate and flue gas vapor
- the sensitivity of metal fate to RKS operating conditions
- the dependence of metal emissions on chlorine content of the incinerated organic waste.

This testing was conducted in conjunction with the development of a metal partitioning model under a U.S EPA contract with EERC Corporation; data from these tests will be used in part to evaluate the predictive capability of the model.

APPROACH

As indicated in Figure 1, the RKS consists of a rotary kiln primary combustion chamber followed by an afterburner. Combustion gases exiting the afterburner are quenched after which they enter a primary air pollution control system. This system consists of a venturi scrubber and packed column scrubber, and is followed by secondary air pollution control consisting of a carbon bed adsorber and a high efficiency particulate filter.

The feed material consisted of synthetic waste formulations mixed into a clay absorbent material. In a four-week series of eight parametric tests, aqueous mixtures were spiked into the solid material, which was then screw-fed to the RKS.

The tests examined the fate of the hazardous constituent trace elements arsenic (As), lead (Pb), cadmium (Cd), chromium (Cr), and barium (Ba). In addition, the non-hazardous elements, copper (Cu), magnesium (Mg), bismuth (Bi), and strontium (Sr) were spiked into the test feed to establish whether their discharge distributions were comparable to those for the hazardous elements.

The feed concentrations of metals are presented in Table 1.

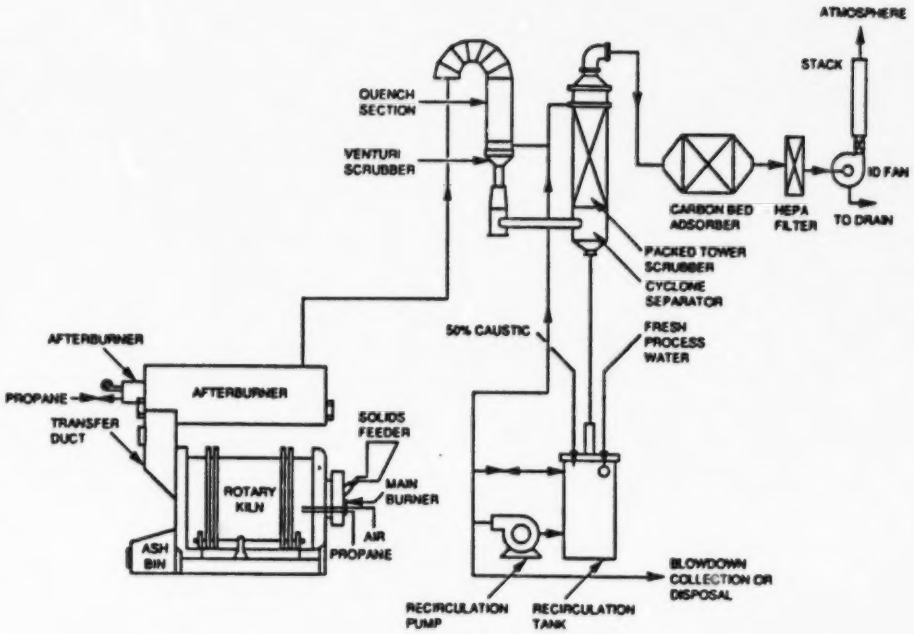

Figure 1. Schematic of the CRF Rotary Kiln System

These values represent the sum of metals in the spike solution and background levels of metals in the feed clay. In addition to the metals, toluene, chlorobenzene, and tetrachloroethylene were added to the solid material in predetermined amounts to achieve the necessary chlorine levels and heat content in the feed.

Kiln temperature, afterburner temperature and the chlorine content of the organic liquid were varied based on a factorial experimental design for three variables over three levels.

The target conditions for the three variables covered the following ranges: feed liquid matrix chlorine content (0 to 8 percent); kiln temperature (816° to 927°C [1500° to 1700°F]); and afterburner temperature (982° to 1204°C [1800° to 2200°F]). Table 2 gives the values for each of the test variables for seven test points specified by the factorial

Table 1. Nominal feed metal concentrations.[a]

Metal	Concentration (ppm)
Arsenic	50
Barium	50
Bismuth	180
Cadmium	10
Chromium	90
Copper	500
Lead	50
Magnesium	17,000
Strontium	300

[a]Based on average clay matrix metals concentrations of: Bi (12 ppm);Cr (53 ppm); Pb (3 ppm); Mg (2.2 percent); Sr (34 ppm)

design algorithm. The eighth test point represents a duplicate of test point 4. This duplicate test condition was added to provide information on test measurement precision.

Table 2. Parametric test design conditions.

Test	Feed Cl Content (wt %)	Kiln Exit Temp, °C (°F)	Afterburner Exit Temp, °C (°F)
1	0	871 (1,600)	1,093 (2,000)
2	4	816 (1,500)	1,093 (2,000)
3	4	927 (1,700)	1,093 (2,000)
4	4	871 (1,600)	1,093 (2,000)
5	4	871 (1,600)	1,204 (2,200)
6	4	871 (1,600)	982 (1,800)
7	8	871 (1,600)	1,093 (2,000)
8[a]	4	871 (1,600)	1,093 (2,000)

[a]Test 8 is a duplicate of test 4 .

Actual test conditions achieved were very close to target test conditions for all test points except point 2. The average afterburner exit temperature was approximately 22°C lower than the target temperature of 1093°C for this test. For all other parametric tests, average actual operating temperatures were within 10°C of target conditions.

Actual feed chlorine content was 0 percent for test 1; 8.3 percent for test 7; and ranged from 3.4 to 4.6 percent for tests 2 through 6, and test 8.

All tests were conducted at the same nominal kiln exit flue gas oxygen (12%), afterburner exit flue gas oxygen (7.5%), and synthetic waste feedrate (63 kg/hr [140 lb/hr]), of which 18 kg/hr (40 lb/hr) was the organic liquid matrix.

For all tests, the kiln rotational speed was held constant at 0.2 rpm, and the primary APCDs were operated at the following nominal settings: scrubber blowdown rate (2 L/min); venturi liquid flowrate (75 L/min); venturi pressure drop (6 kPa); packed tower liquid flowrate (110 L/min).

Sampling for each test included:

- a composite sample of all feed material (clay/organic liquid mixture just before kiln introduction, and aqueous metal spike solution)
- samples of scrubber blowdown water over time
- composite samples of kiln ash from each test
- samples of the flue gas at the afterburner and scrubber exits for metal capture in the particulate and backup impinger train
- samples of the flue gas at the afterburner and scrubber exits for volatile organic hazardous constituents. (2)

RESULTS

Table 3 represents a summary of metal discharge distribution data collected during the eight parametric tests.

Metal Volatility

Metals exiting the kiln followed

Table 3. Normalized metal discharge distributions (% of total measured).

Test Number:	2	4	8	3	6	4	8	5
Primary Variable:	Kiln Exit Temperature (^{o}C)				Afterburner Temperature (^{o}C)			
Target Value:	816	871	871	927	982	1093	1093	1204
Test Average:	825	875	876	927	984	1088	1093	1196
Constants 1;2:	Afterburner=1093^{o}C; Cl=4%				Kiln= 871^{o}C; Cl=4%			
Const. 1 Avg.:	1071	1088	1093	1092	875	878	873	871
Const. 2 Avg.:	3.7	3.8	4.6	4.2	3.4	3.8	4.6	3.6
Arsenic								
Kiln Ash	94.4	86.1	92.3	84.0	93.6	86.1	92.3	91.2
Scrub.Ex. Gas	2.1-2.9	3.8-5.8	2.3-4.1	6.8-8.4	2.6-3.8	3.8-5.8	2.3-4.1	3.0-4.3
Scrub. Water	2.7	8.2	3.6	7.6	2.6	8.2	3.6	4.6
Barium								
Kiln Ash	74.3	79.6	69.9	69.6	85.2	79.6	69.9	86.9
Scrub.Ex. Gas	3.8	2.2	5.5	1.6	2.2	2.2	5.5	1.6
Scrub. Water	21.9	18.2	24.7	28.8	12.5	18.2	24.7	11.5
Bismuth								
Kiln Ash	25.8	22.2	30.0	22.9	20.9	22.2	30.0	30.1
Scrub.Ex. Gas	41.5	41.1	35.2	50.7	47.4	41.1	35.2	37.1
Scrub. Water	32.6	36.7	34.7	26.3	31.6	36.7	34.7	32.8
Cadmium								
Kiln Ash	<15.2	<10.3	<12.9	<10.7	<13.9	<10.3	<12.9	<14.5
Scrub.Ex. Gas	43-49	56-61	42-45	62-69	61-69	56-61	42-45	55-62
Scrub. Water	42-51	34-39	45-55	27-31	25-31	34-39	45-55	31-38
Chromium								
Kiln Ash	94.7	94.1	85.9	95.3	95.5	94.1	85.9	89.3
Scrub.Ex. Gas	3.0	2.0	1.9	2.1	1.1	2.0	1.9	4.2
Scrub. Water	2.3	3.9	12.2	2.6	3.4	3.9	12.2	6.6
Copper								
Kiln Ash	84.2	75.8	76.2	82.3	79.2	75.8	76.2	75.1
Scrub.Ex. Gas	12.9	15.1	17.8	14.1	15.2	15.1	17.8	16.0
Scrub. Water	3.0	9.1	5.9	3.6	5.6	9.1	5.9	8.9
Lead								
Kiln Ash	12.6	15.0	13.7	10.4	5.8	15.0	13.7	13.8
Scrub.Ex. Gas	50.4	48.9	50.2	67.2	60.6	48.9	50.2	45.0
Scrub. Water	37.0	36.1	36.0	22.4	33.6	36.1	36.0	41.1
Magnesium								
Kiln Ash	99.4	99.3	99.3	99.5	99.3	99.3	99.3	99.2
Scrub.Ex. Gas	0.2	0.1	0.2	0.1	0.1	0.1	0.2	0.1
Scrub. Water	0.4	0.6	0.5	0.4	0.6	0.6	0.5	0.7
Strontium								
Kiln Ash	82.9	93.0	89.8	90.1	94.3	93.0	89.8	81.9
Scrub.Ex. Gas	1.1	1.7	3.5	1.6	1.3	1.7	3.5	3.7
Scrub. Water	16.0	5.3	6.6	8.3	4.4	5.3	6.6	14.4

Table 3. (Cont'd).

Test Number:	1	4	8	7
Primary Variable:	Feed Chlorine Content (wt %)			
Target Value:	0	4	4	8
Test Average:	0	3.8	4.6	8.3
Constants 1;2:	Kiln = 871°C; AB = 1093°C			
Const. 1 Avg.:	872	878	873	871
Const. 2 Avg.:	1094	1088	1093	1092
Arsenic				
Kiln Ash	93.9	86.1	92.3	92.4
Scrub.Ex. Gas	1.7-2.2	3.8-5.8	2.3-4.1	4.0-4.8
Scrub. Water	3.9	8.2	3.6	2.7
Barium				
Kiln Ash	68.8	79.6	69.9	78.6
Scrub.Ex. Gas	2.0	2.2	5.5	2.4
Scrub. Water	28.8	18.2	24.7	19.0
Bismuth				
Kiln Ash	64.8	22.2	30.0	36.3
Scrub.Ex. Gas	15.7	41.1	35.2	38.4
Scrub. Water	19.5	36.7	34.7	25.4
Cadmium				
Kiln Ash	<29.3	<10.3	<12.9	<9.3
Scrub.Ex. Gas	42-54	56-61	42-45	68-74
Scrub. Water	29-46	34-39	45-55	68-74
Chromium				
Kiln Ash	95.7	94.1	85.9	92.1
Scrub.Ex. Gas	1.4	2.0	1.9	2.8
Scrub. Water	2.8	3.9	12.2	5.1
Copper				
Kiln Ash	97.6	75.8	76.2	58.0
Scrub.Ex. Gas	0.8	15.1	17.8	33.2
Scrub. Water	1.6	9.1	5.9	8.8
Lead				
Kiln Ash	83.7	15.0	13.7	6.0
Scrub.Ex. Gas	11.6	48.9	50.2	73.6
Scrub. Water	4.7	36.1	36.0	20.3
Magnesium				
Kiln Ash	99.6	99.3	99.3	99.4
Scrub.Ex. Gas	0.03	0.1	0.2	0.1
Scrub. Water	0.36	0.6	0.5	0.5
Strontium				
Kiln Ash	91.8	93.0	89.8	90.7
Scrub.Ex. Gas	2.5	1.7	3.5	1.6
Scrub. Water	5.7	5.3	6.6	7.7

one of three pathways; they either left in the kiln ash, or traveled to the afterburner as fly ash or as volatilized compounds. In this discussion, those metals with a propensity towards partitioning in the kiln ash will be referred to as "refractory" metals, while those which tend to exit from the kiln to the afterburner can be grouped into a category of "volatile" metals. These designations are based on the amount of each metal found in the kiln ash, as a percentage of the total metal measured among the process streams (ash, scrubber blowdown, and flue gas). An assumption in making these distinctions is that metals traveling from the kiln to the afterburner are predominantly the result of volatilization, rather than entrainment in fly ash.

Cadmium, lead, and bismuth appeared to be the most volatile of the nine metals over the range of kiln temperatures tested, with average partitioning to the kiln ash of <15, 20, and 32 percent of the measured amounts for each of the three metals, respectively.

Barium, copper, strontium, arsenic, chromium, and magnesium appeared to be more refractory, with average partitioning of 77, 79, 89, 91, 93, and 99 percent to the kiln ash for each of the metals, respectively.

With the exception of arsenic, the observed order in the degree to which the metals volatilize correlates strongly with that which would be predicted by 'volatility temperatures'(the temperatures at which the vapor pressure of the most volatile principal species of each metal under oxidizing conditions is 10^{-6} atm). The fact that arsenic is significantly less volatile than expected suggests that either a refractory compound is preferred over the predominant arsenic species (As_2O_3), or that some chemical interaction, such

as strong adsorption to the clay, occurred. (4)

Kiln Ash Partitioning

The only metals for which kiln ash partitioning appeared to be affected by changes in kiln temperature were the most volatile metals (cadmium and lead) and arsenic. For these three metals, slight decreases in kiln ash fractions were seen with increasing kiln temperature. Kiln temperature effects for the other individual metals, as well as for total metals, were not significant within data variability.

The effect of increasing feed chlorine content on the partitioning of metals appeared to be much more significant than that of kiln temperature. As kiln and afterburner temperatures were held constant, and feed chlorine content increased from 0.0 to 8.3 percent, the overall fraction of metals partitioning to the kiln ash dropped from 81 to 63 percent.

Chlorine content of the feed affected the volatility of individual metals to different degrees. Kiln ash partitioning of the three volatile metals decreased measurably as chlorine content rose. In the case of bismuth, the fraction of the measured metal found in the kiln ash dropped from 65 to 36 percent as feed chlorine concentration increased from 0.0 to 8.3 percent. Kiln ash fractions of cadmium and lead dropped from 29 to 9 percent, and from 84 to 6 percent, respectively, over the same chlorine range.

Only one of the refractory metals showed an increase in volatility with increasing feed chlorine content. The fraction of measured copper found in the kiln ash dropped from 98 to 58 percent as feed chlorine increased. The volatility of the other refractory metals appeared relatively stable with increasing feed chlorine.

Scrubber Efficiency

The split of metals between scrubber exit gas and scrubber water determines the apparent scrubber efficiency, defined as the ratio of metals in the water to the sum of the metals in the two splits.

Scrubber efficiency for total metals ranged from 33 to 56 percent, and appeared to be negatively impacted by increases in kiln temperature and feed chlorine content.

Average scrubber efficiencies for the individual metals ranged from a low of 32 percent for copper to a high of 88 percent for barium. Average scrubber efficiency for the three volatile metals was lower than that for five of the six refractory metals.

Kiln temperature effects on scrubber efficiency were measurable in the cases of two of the volatile metals. Decreases in scrubber efficiency, from 51 to 31 percent for cadmium and from 42 to 25 percent for lead, were seen with increases in kiln temperature from 825°C to 927°C. This is consistent with the changes in volatilization of these metals experienced with changes in kiln temperature.

The effect of chlorine on scrubber efficiency for individual metals was significant for copper, arsenic, bismuth, and cadmium. In each case, the effect of increasing feed chlorine was to decrease scrubber efficiency. Decreases were as follows: copper (from 67 to 21 percent); arsenic (67 to 38 percent); cadmium (46 to 26 percent); and bismuth (55 to 40 percent).

The effect of afterburner temperature on scrubber efficiency for individual metals was less substantial than that of kiln temper-

ature and feed chlorine content. Modest increases in scrubber efficiency were seen along the increasing afterburner temperature range for bismuth and lead.

Flue Gas Phase Distribution

Standard Method 5 trains were used to collect samples of flue gas particulate- and vapor-phase metals. In this discussion, two assumptions regarding this sampling technique are made: (1) all particulate-phase metals are collected on the filters, and (2) all vapor-phase metals are collected in the impingers; vapor-phase metals do not condense on the filters, nor do they pass through the impingers. (In actuality, the 'vapor-phase' likely includes some water-soluble metal that has wept through the filters and collected in the impingers.)

The distribution of metals in the flue gas favored the particulate phase over vapor. Average particulate phase metals as a fraction of total flue gas metals was highest for the three volatile metals, with lead at 96 percent, cadmium at 90 percent and bismuth at 84 percent. For the refractory metals, the fraction of flue gas metals exiting as particulate ranged from 82 percent for copper to 31 percent for barium.

None of the test variables had a clear effect on the split of total flue gas metals between the two phases. Likewise, effects of test variables on the flue gas phase distribution for the individual metals were not significant within data variability. (4)

CONCLUSIONS

In the subject test series, the trace metals cadmium, lead and bismuth were found to be relatively volatile, while barium, copper, strontium, arsenic, chromium and magnesium were relatively non-volatile.

Average kiln ash partitioning ranged from <15 percent for copper to 99 percent for magnesium. Average scrubber efficiencies for the individual metals ranged from 32 percent for copper to 88 percent for barium.

Both kiln ash partitioning and scrubber efficiency appeared to be impacted negatively by increases in feed chlorine content and, to a lesser extent, increases in kiln temperature.

Average particulate-phase metals as a fraction of total flue gas metals ranged from 31 percent for barium to 96 percent for lead. No clear effects of the three test variables on flue gas phase distributions were apparent.

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